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#### COMMITTEE'S REPORT ON RUTHENIUM FALL-OUT INCIDENT

C. J. Borkowski, J. H. Crawford, R. Livingston, R. H. Ritchie, A. F. Rupp, E. H. Taylor (Chairman)

Formerly issued as an internal report January 25, 1960

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OAK RIDGE NATIONAL LABORATORY
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for the
DEPARTMENT OF ENERGY

# INTRA-LABORATORY CORRESPONDENCE

#### OAK RIDGE NATIONAL LABORATORY

January 25, 1960

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# REPORT OF THE COMMITTEE ON THE RUTHENIUM FALL-OUT INCIDENT

#### INTRODUCTION

Investigations of the fall-out incident of November 11 and November 12 by responsible parties (Health Physics Division and Operations Division personnel) have established beyond reasonable doubt that the incident had its origin in the expulsion of particles, heavily contaminated with ruthenium, which had been detached from the walls of the electric fan housing and ducts in the off-gas system associated with the brick stack. All available evidence indicates that the particles were loosened during maintenance work on the exhaust damper and the bearings of the electric fan and were carried up the stack in two bursts as particulate fall-out when this fan was put back into service. Radiographic and chemical analysis showed the activity to be almost entirely ruthenium (Ru<sup>106</sup>) and its daughter rhodium (Rh<sup>106</sup>) with very little, if any, strontium being present.

This report summarizes the findings and sets forth the conclusions and recommendations of the Committee asked to investigate the incident. In every respect the investigation conducted by the Committee confirms the results of the on-the-spet investigation as mentioned above. A detailed chronology extending to the origin of the maintenance work on the off-gas system follows. This chronology was compiled from interviews with numerous people intimately connected with activities surrounding the incident and including E. J. Witkowski and R. L. Newton of Operations; K. Z. Morgan, J. C. Hart, A. D. Warden, H. H. Abee, R. L. Clark, and T. H. J. Burnett of Health Physics; Shift Supervisor D. Phillips; and numerous others of the survey and analytical groups whose contribution is in the form of data. The accounts of the various individuals were found to complement each other exceedingly well and form the basis of the chronology given below.

An appreciation of the physical layout of the gaseous waste disposal system associated with the brick stack is essential to an understanding of the circumstances leading to the incident. A schematic sketch of the off-gas system is shown in Fig. 1. The first element requiring maintenance was the exhaust damper or valve of the electric fan (indicated by A). The second maintenance problem was one of the bearings of the electric fan itself (indicated by B). Hence on two occasions there was opportunity for mechanical vibration and shock to detach particles on the duct walls of this fan. Not shown on the diagram (Fig. 1) is the stack monitoring arrangement. This consists of a gas stream collected from 40 feet above ground level in the stack and conducted by a small diameter pipe (~1 to 1 1/2" diameter) to a collection filter located near ground level. The filter is normally checked daily. An experimental filter-monitor, checked weekly, has been attached to this same sample line during the past several months.

#### CHRONOLOGY

Sept.-Oct. 1959:

Over a several-week period the damper on the exhaust side of the off-gas electric fan was found not to close tightly and to become progressively worse. Routine repairs were started.

Sat., Oct. 31, 1959:

Repairs were completed on damper. Fan operated 4 hours and then shut down because of bearing failure. The bearing failure was coincidental and marked the end of an unusually long and trouble-free period (years) of operation.

Mon., Nov. 2)
Mon., Nov. 9)

Filters from the experimental stack monitor were removed and tested by the Health Physics Division. These filters continuously sampled the stack gases for one week prior to their removal. Filters for both dates showed large quantities of ruthenium. Almost no ruthenium was found in months of previous sampling up to 10/26. An assay of the filter activity by Reynolds revealed it to be essentially all ruthenium and rhodium with very little strontium (see appendix).

Wed., Nov. 11

Repairs completed on the bearing of the off-gas fan. In the course of repairs the fan housing interior and impeller were partially decontaminated with weak caustic and weak nitric acid. They read about 80~R/hr before cleaning. The impeller and shaft were removed and cleaned again. They read about 5~R/hr after cleaning.

Wed., Nov. 11 4:00 p.m. Electric fan turned off after approximately 30 minutes of initial operation. Following normal procedures, continuous operation of the fan was scheduled to commence with a new work day so that performance of the new bearings could be watched during the first day of operation. This routine start-up for continuous operation was scheduled for 8:15 a.m. of the next day.

Wed., Nov. 11 ~ 4:00 p.m. A Health Physics surveyor in the isotope area found his shoes to be contaminated and immediately afterward found the isotope area to be contaminated. The shift supervisor (J. H. Green) was notified and asked to hold personnel over from the normal work day schedule so that clean-up operations could be started.

Wed., Nov. 11 4:00 to 6:00 p.m. Contamination running up to about 10 mr/hr (G-M counter) was first found throughout the isotope area. (Particles giving substantially higher readings were later found.) Up to 3000 counts/min were found on 1st floor of Bldg. 3037 (office building in isotope area). Throughout this period of time the survey was rapidly extended to try to locate the extent of contamination. The general area, including lawns, around the isotope area and eastward was found contaminated. It was quickly realized that the extent of contamination was widespread and not isolated to the isotope area. The contamination was found to be particulate in nature. Health Physics personnel found their air monitors to be normal. The first reaction was that an old incident had been discovered.

Wed., Nov. 11 6:00 to 12:00 p.m.

Upon finding that the contamination was widespread, supervisory personnel were notified at their homes. Shift Supervisor W. Y. Gissel was notified at his home, and in turn called a number of supervisors. For example, H. H. Abee of Health Physics was notified at 10:30 p.m., E. J. Witkowski of Operations Division at 10:00 p.m. Dave Warden of Health Physics was notified at 11:45 p.m., and he and a number of his survey group were called back to the Laboratory. At Abee's request the stack sampler was checked at 11:00 p.m. and found not to be abnormal. The sticky paper fall-out trays were also checked and found not to be abnormal. (However, a more careful examination the next day showed particulate activity; and subsequently, autoradiography of the trays, the standard monitoring procedure,

showed particulate fall-out. The trays were located in the fringe area of the fall-out, and the particles found were not as radioactive as those found elsewhere). By midnight the first results of a gamma spectrometry study by the Health Physics Division clearly showed the activity to be ruthenium. This study did not rule out the presence of strontium.

Thurs., Nov. 12 12:01 to 8:00 a.m.

Shift Supervisor Don Phillips came on duty at 12:00 midnight and was briefed by J. H. Green. The night was spent in making more detailed surveys and in cleaning operations. Dave Warden and his group arrived at about 12:30 a.m. and at 1:00 a.m. men were sent to selected areas including Bldgs. 4500, 4501, 4505, 3500, the cafeteria, the Jones area (the 4500 construction area), and both main parking areas. The inside of all buildings were found to be clean. Loading platforms were contaminated. Particles reading up to 15 mr/hr were found in the East Parking Lot. (The 4500 construction area was uncontaminated;) A few particles reading up to 1.5 mr/hr were found in the Jones area, but these were very infrequent and represented the extreme eastern limit of the contamination. The cafeteria was uncontaminated, but some activity was found near the covered terrace on the east side of the building. The West Parking Lot was clean (a little activity was found later, but it was felt that it could have been brought in by cars entering the area from the eastern end of the Lab). This established the eastwest limits as being most intense east of the brick stack with gradual attenuation to about zero past the Jones area. A quick check of the north-south limits showed no activity on the hilltop north of Bldg. 4500 and none across the creek to the south. A map showing the contaminated areas is given in Fig. 2. All evidence indicated the source of fall-out to be the brick stack; however, there was a strong feeling that it was an old incident for several reasons: ruthenium was found in stack sampling a few weeks earlier (see Nov. 2 and 11), but an abnormal increase was not found in the sampling that night, air monitors and fallout trays gave negative results, Operations personnel were not aware of any incidents that could lead to fall-out, and activity found on grassed areas appeared to be at the base of the grass near the roots while that found on roads was largely near the General washing of streets and sidewalks started at 4:00 The sidewalks were hosed down. The washing of streets was considerably delayed because of defective equipment and great difficulty was experienced through the night. The equipment in question was the watering truck. It had previously been used with chemical solutions in a contamination clean-up and was not back in good condition. The battery of the pump engine was shorted from spillage during previous use, resulting in delay and difficulty, the hose burst in use, etc. Although it was intended to wash down the East Parking Lot, this could not be achieved. By 8:00 a.m. the extent of contamination was known with considerable detail, it was known to be particles containing ruthenium, and it was strongly suspected to be fall-out from the brick stack. There was still confusion on when the incident could have happened.

Thurs., Nov.12 8:15 a.m. The electric fan was put into operation as scheduled. Ray Newton, foreman of the area, upon realizing that the stack was suspected as the source of contamination, ordered the fan off and the steam fan turned on. It had operated for about 20 to 30 minutes. By about 8:15 a.m. Abee had investigated the degree of contamination on cars in the East Parking Lot in relation to the time they occupied the lot. Cars present up to 3:15 p.m. of Nov. 11 were uncontaminated, while those present up to 4:30 p.m. were contaminated. This established the start of the fall-out in the East Parking Lot at between 3:15 and 4:30 p.m. A similar correlation for the parking lot in front of Bldg. 4500 confirmed this time and indicated the fall-out to be of short duration.

Thurs., Nov. 12 8:15 a.m. to 4:00 p.m.

More extensive surveying and cleaning operations were started. Some areas were roped off. Numbers of people with contaminated shoes were found; leather soled shoes were most difficult to clean. Generally, it was found that the activity was not badly tracked from dirty to clean areas. (It was later realized that tracking of activity into clean buildings did take place in a few instances, but this was in the areas of highest fall-out.) Suddenly, between 9:30 and 10:30 a.m. several groups discovered a new fall-out. Previously cleaned areas and previously cleaned shoes were found to be contaminated. The newly contaminated area was quickly found to be north-west of the brick stack with practically the entire brunt of the fall-out on Bldg. 3025 (Solid State Division) and lesser amounts on a path over Bldg. 2005. The extent of this fall-out is shown in Fig. 3. This immediately confirmed the incident to be acute and almost certainly to be associated with the brick stack. At about 12:00 Noon orders were given to close down as many processing operations as possible that used off-gas facilities of the brick stack. The steam operated fan was closed down, and an auxiliary fan and bypass off-gas connection were put into operation. This bypass system was not equipped with a filter. An order was placed with the shops to have a filter box fabricated on an emergency basis. The Health Physics Division, at the request of Witkowski, moved an experimental moving-filter tape-monitor to the stack and began sampling operations. A normal activity level was found - nothing indicative of the ruthenium fall-out. At about 1:15 p.m. the activity level dropped sharply, coincident with the shutting down of the F3P plant operations. In the morning, at about 10:00 a.m., Division Directors were notified by Health Physics of the fall-out and were advised to take precautions with sensitive equipment, minimize pedestrian traffic, etc. At 2:00 p.m. Health Physics groups were dispatched in cars to survey surrounding roads as far as K-25, Gallaher Gate and White Wing Gate. No contamination was found. Car surveying and washing was started in the East Parking Area. Late in the afternoon Health Physics requested all divisions to survey all shoes and to provide shoe covers to all personnel. At 4:00 p.m. a Laboratory-wide announcement was made requesting all personnel to leave the Laboratory with shoe covers and to remove and discard their shoe covers as they entered their cars. All cars were surveyed as they left the parking area and those found to be contaminated were washed. Several pairs of new shoes were provided at the gates to those people found with contaminated shoes that could not be adequately cleaned (tolerance of 0.6 mr/hr beta activity max. per shoe). Samples of activity from the fall-out and from the off-gas and all ventilating ducts of the stack were collected for assay.

Thurs., Nov. 12 4:00 p.m. Cleaning operations of the Laboratory roads and streets continued. At about 7:00 p.m. assay results obtained by R. R. Rickard of Analytical Division showed the fall-out activity to be identical to that found on the walls of the off-gas system of the stack and different from that collected from the cell ventilating duct work. This constituted the final confirmation that the source of particles was the off-gas system of the brick stackl A chemical analysis revealed almost no strontium. Cleaning and surveying operations continued for the next few days. On November 14 the filter boxes with filters that had been built on an emergency basis were installed in the bypass off-gas duct of the stack. Decontamination operations were started on the steam and electric fans preparatory to dismantling for a complete clean-up. For one week after the fall-out, cars were checked in the parking lots and cleaned when necessary. An adhesive label was placed on the rear window of each car surveyed to prevent duplication of effort. In all, about 30 pairs of new shoes were given out. Urinalyses were started on a group of about 50 people. All assays are not complete, having been interrupted by the plutonium incident, but no significant emounts of ruthenium or strontium have been found. Much of the grassed areas in the fall-out path have been staked on a grid and periodic surveys are being continued at these grid points as well as at other established survey points in the Laboratory. The contamination level in the post-fall-out era appears to have

fallen by about a factor of 3, although this may represent only a small movement of the activity closer to the earth during the first few days. It now appears that further attenuation will be at a much slower rate.

#### GENERAL CONCLUSIONS AND RECOMMENDATIONS

- (1) Under the existing conditions and the accepted philosophy of operation, the incident was capably handled by all persons and groups involved.
- (2) Processes or equipment should not be put into operation without two lines of defense against release of radioactivity.
- (3) Continuity of operations must not prevent the shutting down of a process when maintenance work on the associated containment equipment is indicated by the accumulation of high levels of radioactivity.
- (4) The committee urges the management of the Laboratory to consider very carefully the possibility of closing the Laboratory in certain emergencies, and to try to establish objective criteria for such an action.

With the benefit of hindsight, it is clear that the decision taken in the present case did not jeopardize either health or operability. There is, however, a question in the minds of some of the committee as to whether this step should not have been taken under the circumstances and with the lack of knowledge prevailing shortly after midnight (Thursday A.M.).

#### Notes on Above:

Both (3) and (4) obviously require the setting of levels of radioactivity which will initiate the actions indicated. This is a matter for technical evaluation — the point emphasized here is that the basic philosophy underlying radioactive operations should be re-evaluated.

It is recognized that some unfavorable publicity will attend such shut-downs, but this should be balanced by the favorable effect of a demonstrated concern for safety. The committee is divided in its estimate of the relative weights of the two effects.

#### SPECIFIC SUGGESTIONS

A number of more specific suggestions have been received from various persons and divisions affected by the incident. The committee feels that they are sound and should be implemented.

- (A) Pertaining to the off-gas system
  - (1) High levels of radioactivity must not be allowed to accumulate downstream from the final filters.
  - (2) Any process contributing appreciable radioactivity to the system should include equipment for removing it from the off-gas.

- (3) A similar provision should apply to any substance (acid fumes, NH<sub>3</sub>, dust, etc.) which will affect the final purification system adversely.
- (4) All off-gas lines entering the stack should have parallel alternates of equal capability, so connected that repairs or replacements to one do not disturb the other.
- (5) Off-gas operations (and probably other types of waste) should be instrumented for flow and radioactivity with recording at a central control point, to facilitate remedial actions in an emergency.
- (B) Pertaining to radioactive monitoring
  - (1) Improved means for early detection of airborne particulate radioactivity should be sought.
  - (2) Availability of monitoring equipment should be improved.
    - (a) For routine monitoring before leaving (compulsory)

3019 Pilot Plant 3026 Chemical Separation Laboratory 3508 Isolation Laboratory

(b) Friskers for gross amounts of activity

Fast and West Portals 2010 Cafeteria

(c) Available for use when needed

4500 Central Research Laboratory

- (3) Improvements in monitoring available with little or no development.
  - (a) Continuously recording, 3 x 3-inch NaI scintillation counters monitoring the general background at a number of positions could have detected the rise in activity caused by the Ru incident.
  - (b) Several methods are available for improving the reliability of personnel monitors (friskers) to reduce the number of spurious alarms. These include such procedures as gating the circuits with an electric eye, measuring rise-time (from movement of contaminated person past counter) rather than level alone, etc.
- (4) The committee notes that a more serious release of radioactivity over a larger area might make an aerial survey very useful. Since this can be done on short notice (requiring only getting equipment and a man to the airport), the procedure for doing it should be incorporated into emergency plans, and made known to potential emergency directors.

- (C) Pertaining to emergency procedures
  - (1) The central emergency warning station should have a list of areas sensitive to contamination (counting rooms, etc.). As soon as possible, consistent with human safety and protection of major facilities, these sensitive areas should be warned of any release of radioactivity.
  - (2) Communications with personnel present during an incident should be faster and more complete than at present, again consistent with overriding local considerations.
    - (a) The public address system is available, and practice in its use may be invaluable preparation for a more serious occurrence.
    - (b) The plans for handling a continuing abnormal situation of this sort should include a central clearing house for information separate from the Shift Supervisors' phone. It could be an unlisted number known only to divisional offices. This would require a clerk or secretary for the Shift Supervisors.
    - (c) The bulletin issued at the Portals during the plutonium incident is highly commended.
- (D) Pertaining to other aspects of laboratory operation
  - (1) The day shift should put emergency equipment (water truck, hoses, etc.) into working order before leaving. There are not enough men on the night shift to make repairs. The failure of the water truck, which had been used for a job during the day, prevented the cleaning of the parking lot during the night of Wednesday-Thursday.
  - (2) Since rubber-soled shoes proved much easier to decontaminate than leather, attention should be given to the type of sole used on safety shoes. It is thought that Neolite is presently used. It probably resembles rubber in decontaminability.

Committee on the Ruthenium Fall-Out Incident,

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# Appendix: Technical Aspects of Fallout Incident November 11-12, 1959

A detailed report on the nature and extent of the fallout is not possible at the present time. The following preliminary considerations may be revised in the light of later findings.

#### Particle Size and Composition

Early evidence pointed to contamination in the form of sizeable radioparticulates, e.g., 1) the continuous air monitors in the area showed no increase in activity, indicating that the particles were so large that they had low probability of being swept into the sampling air stream; 2) the twenty-four hour stack samples likewise indicated no unusual increase in activity; and 3) the narrow and fairly well-defined area of fallout indicated that the particles settled at a rapid rate.

Subsequent work by H. H. Abee, W. D. Cottrell and associates has resulted in the isolation of a few large particles. One such particle, which gave a survey meter reading of 20 mr/hr at contact, was clearly visible to the naked eye and had a diameter of about 1 mm. It was translucent and had a crystalline appearance, not at all suggestive of iron scale or rust. X-ray diffraction studies, although very preliminary, seem to indicate that the particles examined are neither ruthenium metal nor ruthenium oxide.

An approximate idea of the average particle size in the East parking lot may be had by assuming that the particles settled according to Stokes' law after leaving the top of the stack. Using a 7 m.p.h. wind as recorded at 4:00 p.m. on November 11 and an effective stack height of 260 ft, one finds that the settling velocity would have been 1.67 ft/sec for this case. Assuming a density of 1.0 gm/cm<sup>3</sup> for the particle, one finds a diameter of 0.13 mm. Such a particle would be classed with heavy industrial dusts and is an order of magnitude larger than the minimum size visible to the unaided eye.

#### Estimate of Total Fallout

The data on which to base an estimate of the total quantity of Ru deposited in the area are rather meagre. The G.M. survey meter readings indicate mainly the level of incident beta radiation but are difficult to interpret quantitatively because of a strong variation in counter sensitivity with electron energy.

Some readings were taken in the contaminated area on the evening of November 13th with a scintillation counter survey meter which responds mainly to gamma radiation. These readings were much less sensitive to height above ground, and distance from the fallout zone than were the G.M. survey meter readings. A number of traverses of the contaminated area indicated that in the neighborhood of the stack

the gamma radiation level was approximately 0.4 mr/hr while in the grassy area in front of the 4500 Building the reading was about .05 mr/hr at the extreme west end, and approximately .01 mr/hr close to the East parking lot. All readings were taken at approximately three feet above ground level.

To interpret these readings in terms of the Ru activity it is necessary to estimate the dose response of the instrument to the incident photons. Assuming an average gamma energy of 0.56 MeV, and using the approximate relation between dose D (mr/hr) and E (photon energy in MeV)

$$D = \frac{5.6 \times 10^2}{E}$$

one finds that a photon flux of  $10^3$  photons/cm<sup>2</sup> sec from the disintegration of  $\rm Rh^{106}$  is equivalent to a dose of 1 mr/hr. An experimental calibration of the instrument with a ruthenium source of known activity gave good agreement with this estimate.

A rough: idea of the activity deposited in the neighborhood of the stack may be had if one assumes that the gamma reading of 0.4 mr/hr is due to a uniform disc source of Ru of 300' radius centered about the stack. If  $\sigma$  is the superficial density of this source in photons/cm<sup>2</sup> and if  $\ell$  is the attenuation length of 0.56 Mev photons

in air ( $\ell \sim 550^{\circ}$ ), then the photon flux  $\phi$  at height h above the center of such a source is given by

$$\Phi = \frac{\sigma}{2} \left\{ E_1(h/\ell) - E_1 \left[ (1/\ell) \sqrt{h^2 + y_0^2} \right] \right\}$$

where  $y_0 = 300^\circ$ , the disc radius, and  $E_1(x) = \int_0^\infty e^{-xy} \, dy/y$ . One may calculate  $\sigma$  from this equation if one sets  $\bullet = (0.4 \text{ mr/hr}) \times 10^3$  photons/cm<sup>2</sup> sec/mr hr<sup>-1</sup>. The total activity turns out to be  $\sim 3$  curies if one assumes 1/3 photon per disintegration of Ru<sup>106</sup>. Similar rough considerations yield a value of  $\sim 0.3$  curie for the total activity deposited in the area east of Fifth Street.

It should be noted that these estimates are probably on the low side because 1) the gamma measurements were made after considerable washing of streets and sidewalks had occurred, and 2) it is assumed that gamma-ray attenuation by irregularities in terrain, buildings, trees, etc., was unimportant.

Estimates based on observation of the distribution in depth of 0.1 - 0.2 mm particles deposited at random in grassed areas indicates that the above values may possibly be low by a factor of 2. If one estimates terrain effects and attenuation by objects in the contaminated area one finds an upper limit of ~ 5 times the values given above, i.e., ~ 15 curies in the area around the stack and ~ 1.5 curies in the area east of Fifth Street.

#### Residual Activity

Figure 4 shows a survey map of the Laboratory area with radiation levels before and after the fallout (October 29th and November 18th) indicated at various points. These values were obtained by J. S. Sprain of the Health Physics Division in the regular monthly area survey program. Readings were taken with a G.M. survey meter at a height of ~3 ft above the road surface from a fixed position at the rear of a panel truck. The counter is calibrated in units of mr/hr with a radium source. The mr/hr readings given in Figure 4 should be regarded as nominal only since an unknown fraction of the reading is due to beta radiation.

This map shows that clean-up operations on roads and sidewalks have been effective in reducing radiation levels to values approaching those which existed before the accident, with a few exceptions.

There seems to have been negligible contamination of low-level counting areas throughout the Laboratory. The X-ray laboratory in Building 3025 became contaminated to some extent on November 12th, but has been thoroughly cleaned and at present the background is no greater than it was before the accident.

#### Ruthenium Levels in Experimental Stack Monitoring Equipment

It is interesting to note that some warning of the impending fallout would have been available before the accident if stack monitoring equipment of the sort used by J. W. Youngblood had been employed in a routine short-time sampling procedure. His experimental apparatus is being employed at present to determine the kinds of activity which are released into the stack and to test the efficiency of various methods of removal. The particulate samples are taken from the same line used for the regular twenty-four hour samples but are collected over a one-week period. After collection, the samples are analyzed by Sam Reynolds' group and the results of analysis are made available to Youngblood approximately one week after they are submitted. The following air activities were found assuming a constant rate of release of Ru 106 over the periods indicated.

Sampling Period	106	sr <sup>90</sup>
10/7 - 10/14	1.9 × 10 <sup>-11</sup> μc/cm <sup>3</sup>	$0.6 \times 10^{-10}  \mu c/cm^3$
10/14 - 10/26	1.3 × 10 <sup>-11</sup>	0.3 × 10 <sup>-10</sup>
10/26 - 11/2	2.2 × 10 <sup>-7</sup>	2.3 × 10 <sup>-10</sup>
11/2 - 11/9	$1.4 \times 10^{-7}$	6.5 × 10 <sup>-10</sup>
11/9 - 11/18	1.2 × 10 <sup>-8</sup>	1.6 × 10 <sup>-10</sup>
7/31 - 8/7	$1.3 \times 10^{-10}$	$8.2 \times 10^{-10}$

There seems to be little correlation between Sr<sup>90</sup> and Ru<sup>106</sup> activities found in the several months in which this apparatus has been operative. As has been noted above, the routine twenty-four hour stack sample showed no marked increase in gross counting rate during these periods.

One is hard put to explain the fact that about twenty times as much activity was observed during the week of the damper repair as during the period in which the actual fallout was supposed to have occurred. A possible explanation is that the Ru<sup>106</sup> released between October 26th and November 9th was in the form of very small particles and was carried great distances from the stack before being deposited.

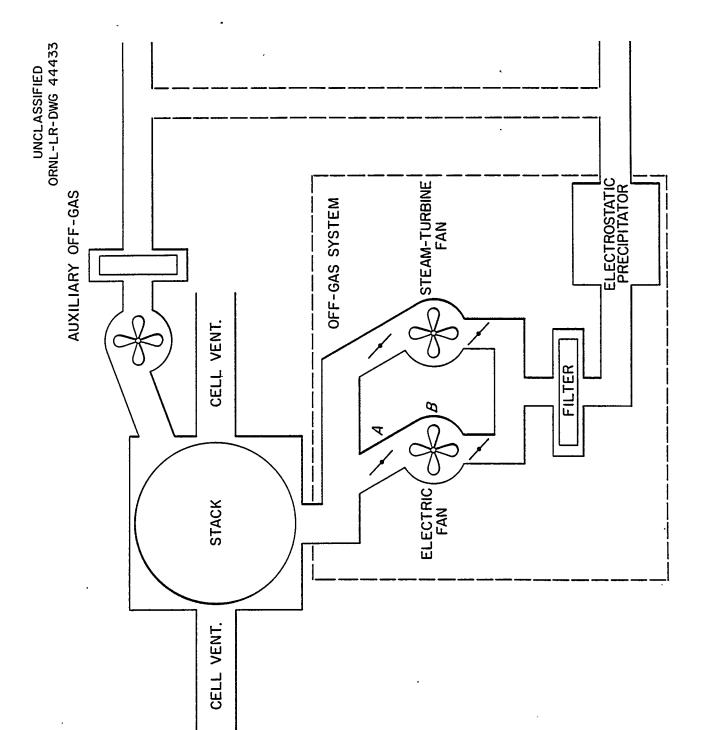
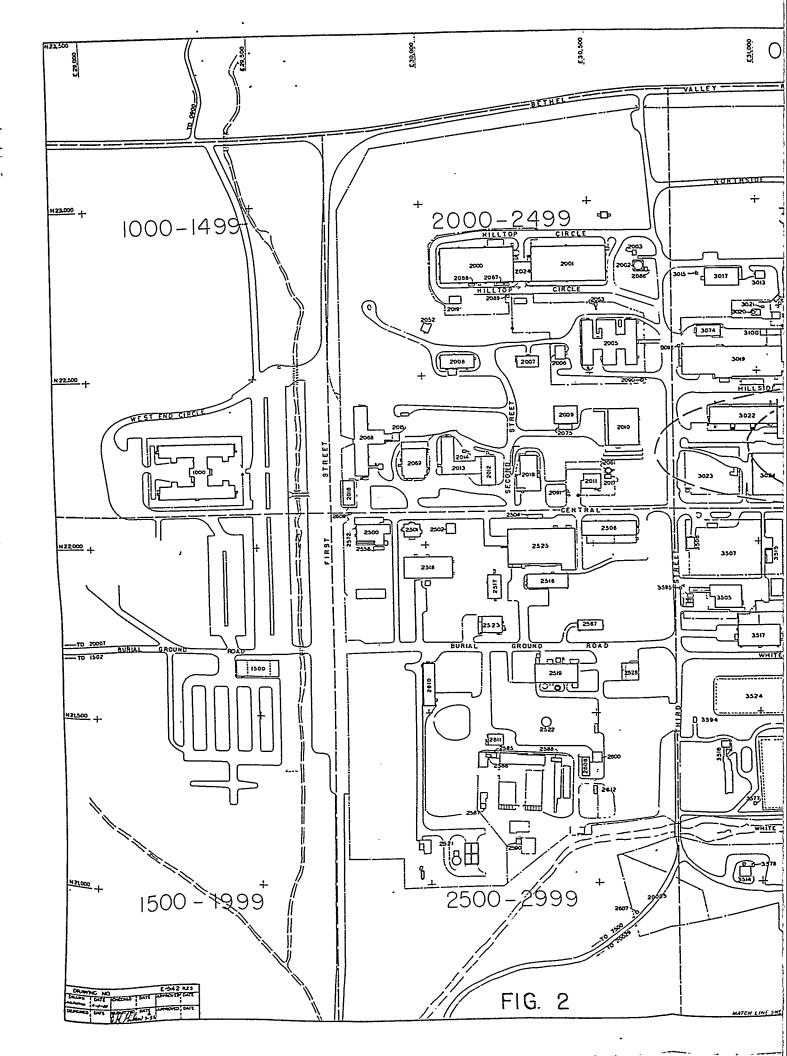
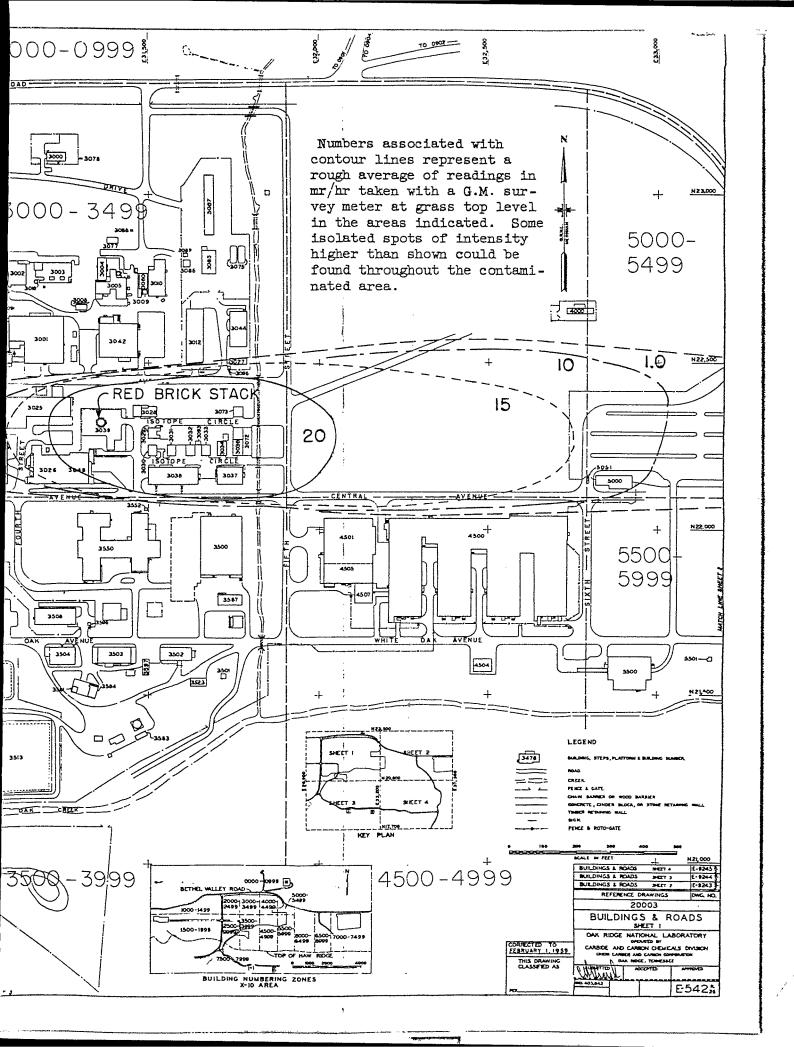
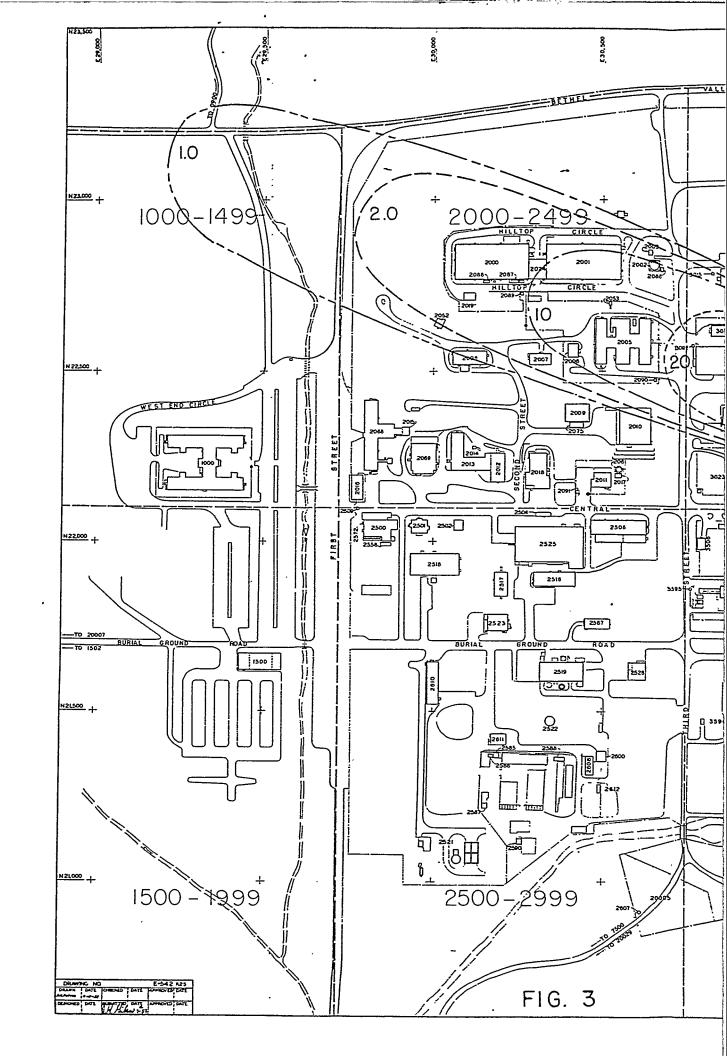
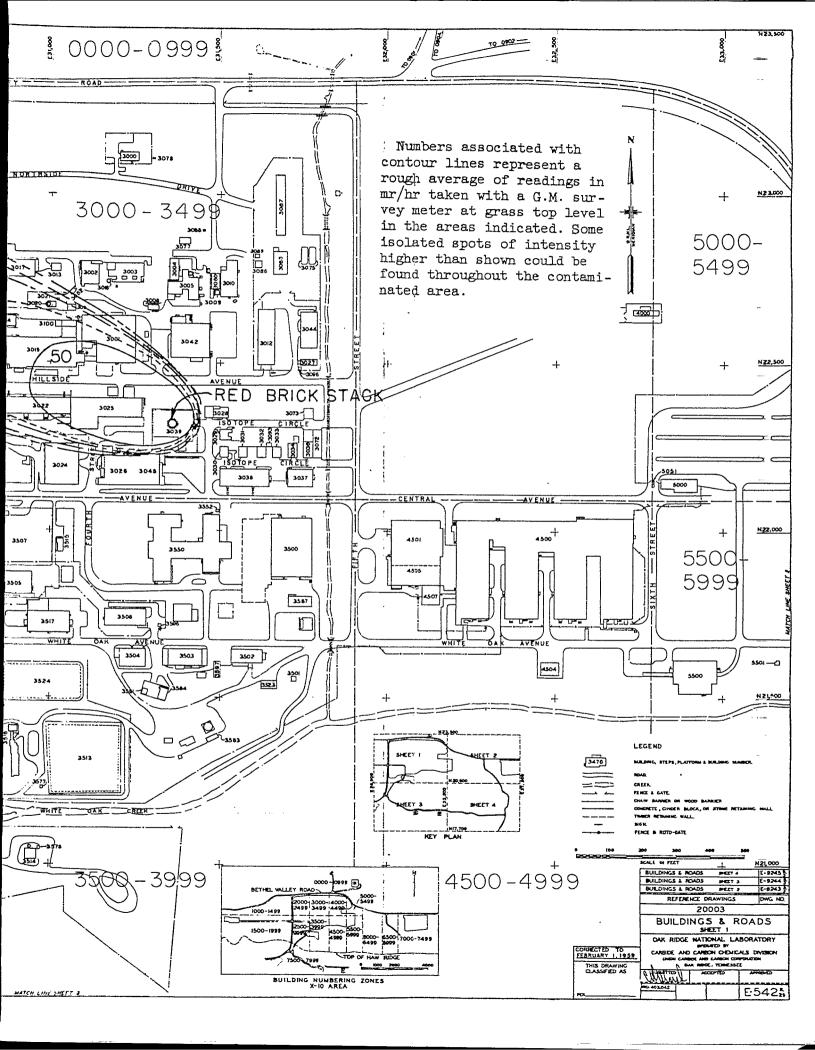


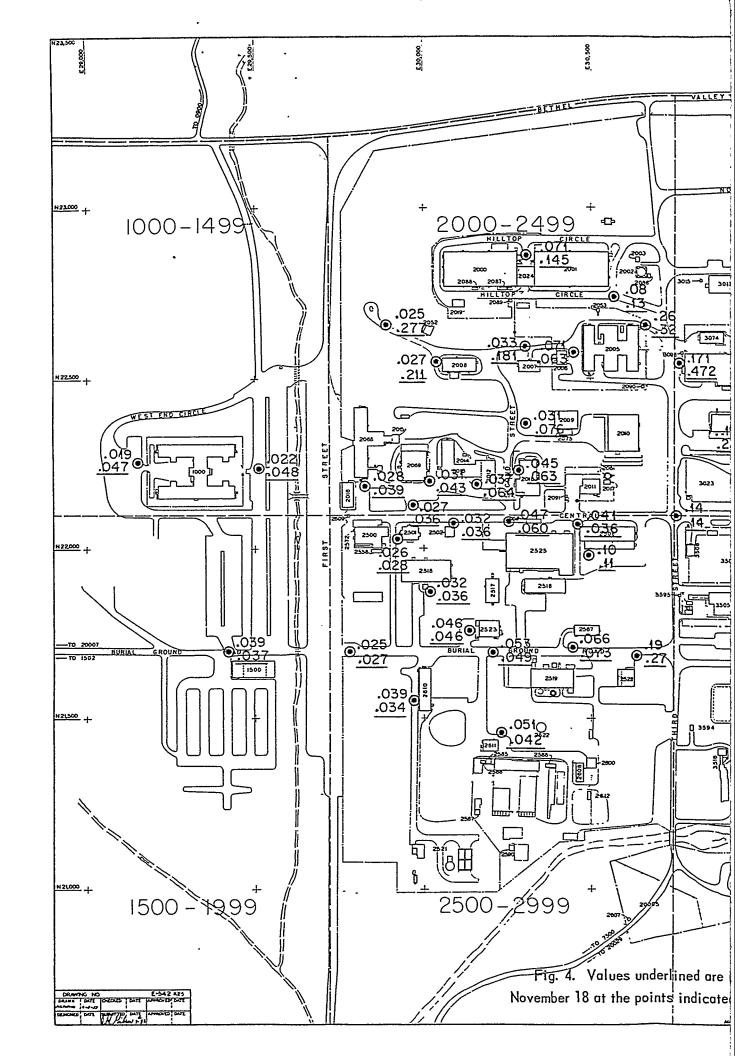
Fig.

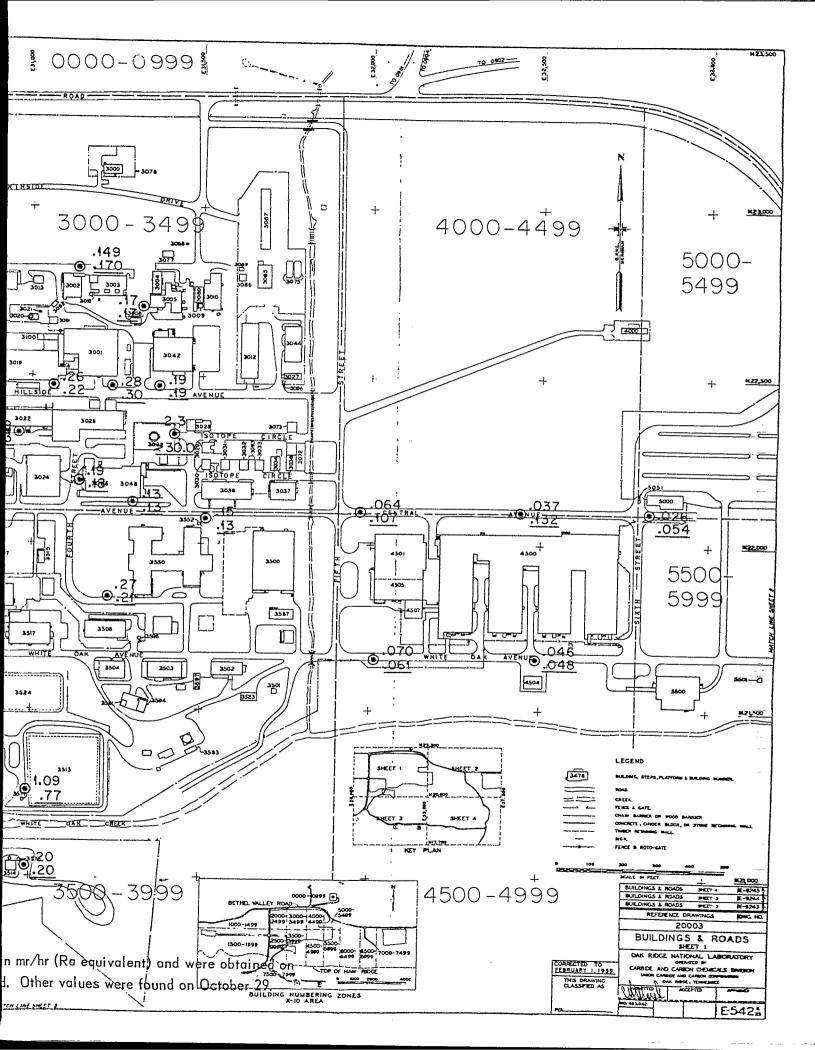












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